

08-23-00

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UTILITY PATENT APPLICATION TRANSMITTAL
(Large Entity)*(Only for new nonprovisional applications under 37 CFR 1.53(b))*Docket No.
LT1101Total Pages in this Submission
60**TO THE ASSISTANT COMMISSIONER FOR PATENTS**Box Patent Application
Washington, D.C. 20231

Transmitted herewith for filing under 35 U.S.C. 111(a) and 37 C.F.R. 1.53(b) is a new utility patent application for an invention entitled:

POLY(ARYLENE ETHER) ADHESIVE COMPOSITIONS

and invented by:

Michael J. Davis et al.08/22/00
U.S. PTO
09/644012If a **CONTINUATION APPLICATION**, check appropriate box and supply the requisite information:☐ Continuation ☐ Divisional ☐ Continuation-in-part (CIP) of prior application No.: _____

Which is a:

☐ Continuation ☐ Divisional ☐ Continuation-in-part (CIP) of prior application No.: _____

Which is a:

☐ Continuation ☐ Divisional ☐ Continuation-in-part (CIP) of prior application No.: _____

Enclosed are:

Application Elements

1. ☒ Filing fee as calculated and transmitted as described below
2. ☒ Specification having 32 pages and including the following:
 - a. ☐ Descriptive Title of the Invention
 - b. ☒ Cross References to Related Applications *(if applicable)*
 - c. ☐ Statement Regarding Federally-sponsored Research/Development *(if applicable)*
 - d. ☐ Reference to Microfiche Appendix *(if applicable)*
 - e. ☒ Background of the Invention
 - f. ☒ Brief Summary of the Invention
 - g. ☐ Brief Description of the Drawings *(if drawings filed)*
 - h. ☒ Detailed Description
 - i. ☒ Claim(s) as Classified Below
 - j. ☒ Abstract of the Disclosure

UTILITY PATENT APPLICATION TRANSMITTAL
(Large Entity)

(Only for new nonprovisional applications under 37 CFR 1.53(b))

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LT1101

Total Pages in this Submission

60

Application Elements (Continued)

3. ☐ Drawing(s) *(when necessary as prescribed by 35 USC 113)*
- a. ☐ Formal Number of Sheets _____
- b. ☐ Informal Number of Sheets _____
4. ☒ Oath or Declaration
- a. ☒ Newly executed *(original or copy)* ☐ Unexecuted
- b. ☐ Copy from a prior application (37 CFR 1.63(d)) *(for continuation/divisional application only)*
- c. ☒ With Power of Attorney ☐ Without Power of Attorney
- d. ☐ DELETION OF INVENTOR(S)
Signed statement attached deleting inventor(s) named in the prior application,
see 37 C.F.R. 1.63(d)(2) and 1.33(b).
5. ☐ Incorporation By Reference *(usable if Box 4b is checked)*
The entire disclosure of the prior application, from which a copy of the oath or declaration is supplied under
Box 4b, is considered as being part of the disclosure of the accompanying application and is hereby
incorporated by reference therein.
6. ☐ Computer Program in Microfiche *(Appendix)*
7. ☐ Nucleotide and/or Amino Acid Sequence Submission *(if applicable, all must be included)*
- a. ☐ Paper Copy
- b. ☐ Computer Readable Copy *(identical to computer copy)*
- c. ☐ Statement Verifying Identical Paper and Computer Readable Copy

Accompanying Application Parts

8. ☒ Assignment Papers *(cover sheet & document(s))*
9. ☐ 37 CFR 3.73(B) Statement *(when there is an assignee)*
10. ☐ English Translation Document *(if applicable)*
11. ☐ Information Disclosure Statement/PTO-1449 ☐ Copies of IDS Citations
12. ☐ Preliminary Amendment
13. ☒ Acknowledgment postcard
14. ☒ Certificate of Mailing
- ☐ First Class ☒ Express Mail *(Specify Label No.):* EL480563406US

UTILITY PATENT APPLICATION TRANSMITTAL (Large Entity)

(Only for new nonprovisional applications under 37 CFR 1.53(b))

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LT1101

Total Pages in this Submission
60

Accompanying Application Parts (Continued)

15. ☐ Certified Copy of Priority Document(s) (if foreign priority is claimed)

16. ☐ Additional Enclosures (please identify below):

Fee Calculation and Transmittal

CLAIMS AS FILED

For	#Filed	#Allowed	#Extra	Rate	Fee
Total Claims	36	- 20 =	16	x \$18.00	\$288.00
Indep. Claims	6	- 3 =	3	x \$78.00	\$234.00
Multiple Dependent Claims (check if applicable) <input type="checkbox"/>					\$0.00
BASIC FEE					\$690.00
OTHER FEE (specify purpose)					\$0.00
TOTAL FILING FEE					\$1,212.00

- ☐ A check in the amount of _____ to cover the filing fee is enclosed.
- ☒ The Commissioner is hereby authorized to charge and credit Deposit Account No. 07-0893 as described below. A duplicate copy of this sheet is enclosed.
- ☒ Charge the amount of \$1,212.00 as filing fee.
 - ☒ Credit any overpayment.
 - ☒ Charge any additional filing fees required under 37 C.F.R. 1.16 and 1.17.
 - ☐ Charge the issue fee set in 37 C.F.R. 1.18 at the mailing of the Notice of Allowance, pursuant to 37 C.F.R. 1.311(b).


Signature

J. Michael Buchanan
Reg. No. 44, 571

Dated: August 22, 2000



23413

PATENT TRADEMARK OFFICE

Cantor Colburn LLP
55 Griffin Road South
Bloomfield, CT 06002

CC:

ASSIGNMENT

In consideration of One Dollar and other good and valuable consideration, of which we acknowledge receipt, we, Michael J. Davis, 624 Ridgewood Drive, Coshocton, OH 43812; James Estel Tracy, P.O. Box 95 - 225 Allison Drive, Killbuck, OH 44637; Geoffrey Henry Riding, 21 Bellwood Lane, Castleton, NY 12033; and Gary William Yeager, 2010 Fairlawn Parkway, Schenectady, NY 12309, sell and assign to General Electric Company, a New York Corporation, its successors and assigns the entire right, title and interest in and to the improvements in POLY(ARYLENE ETHER) ADHESIVE COMPOSITIONS invented by us, as described in the application for United States Patent executed concurrently herewith, and any and all applications for patent and patents therefor in any and all countries, including all divisions, reissues, continuations and extensions thereof, and all rights of priority resulting from the filing of said United States application, and authorize and request any official whose duty it is to issue patents, to issue any patent on said improvements or resulting therefrom to said General Electric Company, or its successors or assigns and agree that on request and without further consideration, but at the expense of said Company, we will communicate to said Company or its representatives or nominees any facts known to me respecting said improvements and testify in any legal proceeding, sign all lawful papers, execute all divisional, continuing and reissue applications, make all rightful oaths and generally do everything possible to aid said Company, its successors, assigns, and nominees to obtain and enforce proper patent protection for said improvements in all countries.

_____, 2000
Date

Michael J. Davis

STATE OF)
) ss at:
COUNTY OF)

This ____ day of _____, 2000, before me personally came the above-named, to me personally known as the individual who executed the foregoing assignment, who acknowledged to me that executed the same of own free will for the purposes therein set forth.

Notary Public
My Commission Expires: _____

Notary Public
My Commission Expires: _____

2000

Harry William Loeger

STATE OF

)

) ss at:

COUNTY OF

)

Susan M. Zampieri

~~Notary Public~~

My Commission Expires: 11/11/00

SUSAN ZAMPIER

Notary Public in the State of New York

Qualified in Saratoga Co.

No. 01ZA5068047

My Commission Expires Nov. 11, 2000

ASSIGNMENT

In consideration of One Dollar and other good and valuable consideration, of which we acknowledge receipt, we, Michael J. Davis, 624 Ridgewood Drive, Coshocton, OH 43812; James Estel Tracy, P.O. Box 95 - 225 Allison Drive, Killbuck, OH 44637; Geoffrey Henry Riding, 21 Bellwood Lane, Castleton, NY 12033; and Gary William Yeager, 2010 Fairlawn Parkway, Schenectady, NY 12309, sell and assign to General Electric Company, a New York Corporation, its successors and assigns the entire right, title and interest in and to the improvements in POLY(ARYLENE ETHER) ADHESIVE COMPOSITIONS invented by us, as described in the application for United States Patent executed concurrently herewith, and any and all applications for patent and patents therefor in any and all countries, including all divisions, reissues, continuations and extensions thereof, and all rights of priority resulting from the filing of said United States application, and authorize and request any official whose duty it is to issue patents, to issue any patent on said improvements or resulting therefrom to said General Electric Company, or its successors or assigns and agree that on request and without further consideration, but at the expense of said Company, we will communicate to said Company or its representatives or nominees any facts known to me respecting said improvements and testify in any legal proceeding, sign all lawful papers, execute all divisional, continuing and reissue applications, make all rightful oaths and generally do everything possible to aid said Company, its successors, assigns, and nominees to obtain and enforce proper patent protection for said improvements in all countries.

June 28, 2000
Date

Michael J. Davis
Michael J. Davis

STATE OF OH)
) ss at:
COUNTY OF Coshocton)

This 28th day of June, 2000, before me personally came the above-named, to me personally known as the individual who executed the foregoing assignment, who acknowledged to me that executed the same of own free will for the purposes therein set forth.

Patricia Robinson
Notary Public
My Commission Expires: 11/5/2004

James Estel Tracy

COUNTY OF) ss at:
)

Notary Public
My Commission Expires: _____

Geoffrey Henry Riding

STATE OF)
) ss at:
COUNTY OF)

Notary Public
My Commission Expires: _____

STATE OF)
) ss at:
COUNTY OF)

Notary Public
My Commission Expires: _____

ASSIGNMENT

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_____, 2000

Date

Michael J. Davis

STATE OF)
) ss at:
COUNTY OF)

This ____ day of _____, 2000, before me personally came the above-named, to me personally known as the individual who executed the foregoing assignment, who acknowledged to me that executed the same of own free will for the purposes therein set forth.

Notary Public
My Commission Expires: _____

June 28, 2000
Date

James Estel Tracy
James Estel Tracy

STATE OF OH

COUNTY OF) ss at:
Coshocton)

This 28th day of June, 2000, before me personally came the above-named, to me personally known as the individual who executed the foregoing assignment, who acknowledged to me that executed the same of own free will for the purposes therein set forth.

Leticia A. Robinson
Notary Public
My Commission Expires: 11/5/2004

_____, 2000
Date

Geoffrey Henry Riding

STATE OF)
COUNTY OF) ss at:
)

This ____ day of _____, 2000, before me personally came the above-named, to me personally known as the individual who executed the foregoing assignment, who acknowledged to me that executed the same of own free will for the purposes therein set forth.

Notary Public
My Commission Expires: _____

002230"27044960

Gary William Yeager

This ____ day of _____, 2000, before me personally came the above-named, to me personally known as the individual who executed the foregoing assignment, who acknowledged to me that executed the same of own free will for the purposes therein set forth.

Notary Public
My Commission Expires: _____

ASSIGNMENT

In consideration of One Dollar and other good and valuable consideration, of which we acknowledge receipt, we, Michael J. Davis, 624 Ridgewood Drive, Coshocton, OH 43812; James Estel Tracy, P.O. Box 95 - 225 Allison Drive, Killbuck, OH 44637; Geoffrey Henry Riding, 21 Bellwood Lane, Castleton, NY 12033; and Gary William Yeager, 2010 Fairlawn Parkway, Schenectady, NY 12309, sell and assign to General Electric Company, a New York Corporation, its successors and assigns the entire right, title and interest in and to the improvements in POLY(ARYLENE ETHER) ADHESIVE COMPOSITIONS invented by us, as described in the application for United States Patent executed concurrently herewith, and any and all applications for patent and patents therefor in any and all countries, including all divisions, reissues, continuations and extensions thereof, and all rights of priority resulting from the filing of said United States application, and authorize and request any official whose duty it is to issue patents, to issue any patent on said improvements or resulting therefrom to said General Electric Company, or its successors or assigns and agree that on request and without further consideration, but at the expense of said Company, we will communicate to said Company or its representatives or nominees any facts known to me respecting said improvements and testify in any legal proceeding, sign all lawful papers, execute all divisional, continuing and reissue applications, make all rightful oaths and generally do everything possible to aid said Company, its successors, assigns, and nominees to obtain and enforce proper patent protection for said improvements in all countries.

_____, 2000
Date

Michael J. Davis

STATE OF)
) ss at:
COUNTY OF)

This ____ day of _____, 2000, before me personally came the above-named, to me personally known as the individual who executed the foregoing assignment, who acknowledged to me that executed the same of own free will for the purposes therein set forth.

Notary Public
My Commission Expires: _____

002230 "2104350

_____, 2000
Date James Estel Tracy

STATE OF

COUNTY OF) ss at:
)

This ____ day of _____, 2000, before me personally came the above-named, to me personally known as the individual who executed the foregoing assignment, who acknowledged to me that executed the same of own free will for the purposes therein set forth.

Notary Public
My Commission Expires: _____

7/13 / 2000
Date

Geoffrey H Riding
Geoffrey Henry Riding

STATE OF New York
) ss at:
COUNTY OF Albany

This 13th day of July, 2000, before me personally came the above-named, to me personally known as the individual who executed the foregoing assignment, who acknowledged to me that executed the same of own free will for the purposes therein set forth.

RALENE L. SMITH
Notary Public, State of New York
No. 01 SM 4951963
Qualified in Albany County
Commission Expires 6/5/01

Ralene L Smith
Notary Public
My Commission Expires: 6/5/01

Gary William Yeager

STATE OF)
)
COUNTY OF)

This ____ day of _____, 2000, before me personally came the above-named, to me personally known as the individual who executed the foregoing assignment, who acknowledged to me that executed the same of own free will for the purposes therein set forth.

Notary Public
My Commission Expires: _____

Study	Year	Sample Size	Age Range	Gender	Intervention	Control	Outcome
1	1998	100	18-25	F
2	2001	150	26-35	M
3	2003	200	36-45	F
4	2005	250	46-55	M
5	2007	300	56-65	F
6	2009	350	66-75	M
7	2011	400	76-85	F
8	2013	450	86-95	M
9	2015	500	96-105	F
10	2017	550	106-115	M
11	2019	600	116-125	F
12	2021	650	126-135	M
13	2023	700	136-145	F
14	2025	750	146-155	M
15	2027	800	156-165	F
16	2029	850	166-175	M
17	2031	900	176-185	F
18	2033	950	186-195	M
19	2035	1000	196-205	F
20	2037	1050	206-215	M
21	2039	1100	216-225	F
22	2041	1150	226-235	M
23	2043	1200	236-245	F
24	2045	1250	246-255	M
25	2047	1300	256-265	F
26	2049	1350	266-275	M
27	2051	1400	276-285	F
28	2053	1450	286-295	M
29	2055	1500	296-305	F
30	2057	1550	306-315	M
31	2059	1600	316-325	F
32	2061	1650	326-335	M
33	2063	1700	336-345	F
34	2065	1750	346-355	M
35	2067	1800	356-365	F
36	2069	1850	366-375	M
37	2071	1900	376-385	F
38	2073	1950	386-395	M
39	2075	2000	396-405	F
40	2077	2050	406-415	M
41	2079	2100	416-425	F
42	2081	2150	426-435	M
43	2083	2200	436-445	F
44	2085	2250	446-455	M
45	2087	2300	456-465	F
46	2089	2350	466-475	M
47	2091	2400	476-485	F
48	2093	2450	486-495	M
49	2095	2500	496-505	F
50	2097	2550	506-515	M
51	2099	2600	516-525	F
52	2101	2650	526-535	M
53	2103	2700	536-545	F
54	2105	2750	546-555	M
55	2107	2800	556-565	F
56	2109	2850	566-575	M
57	2111	290					

POLY(ARYLENE ETHER) ADHESIVE COMPOSITIONS

CROSS REFERENCE TO RELATED APPLICATIONS

This application claims the benefit of U.S. Provisional Application Serial No. 60/212752, filed June 20, 2000.

BACKGROUND OF THE INVENTION

The present invention relates to adhesive compositions for thermoplastic substrates and a method of manufacture thereof. More particularly, the present invention relates to such adhesive compositions comprising poly(arylene ether) polymers.

5 There remains a continuing need in the art for thermosetting adhesives for use with thermoplastic adherends, particularly substrates used in, and associated with, fabricating printed circuit boards. The need for miniaturization of circuit boards has increased and methods such as incorporation of microvias in printed circuit boards have been developed.
10 Furthermore, it would be beneficial to have substrates capable of microvia technology and capable of being laser drilled without interference of reinforcements such as woven or chopped fiberglass.

In the fabrication of printed circuit boards, it is known to use copper foil coated with adhesive, or copper foil with a cured adhesive which is then
15 coated with adhesive and partially cured as described in U.S. Patent No. 5,557,843. This method is limited by the thickness of the copper foil that can be effectively coated and handled for practical use in the printed circuit board or rigid-flex board. Thermoplastic films are known which are metallized with copper or other conductive material to low thicknesses such as 3 microns or 5
20 microns. Examples of metallized polyimides are described in, for example, U.S. Patent Nos. 4,842,946 and 4,959,121. These metallized films can be coated with adhesive and used, for example, in microvia and rigid-flex applications.

Adhesives suitable for bonding thermoplastic adherends, and circuit boards in particular, need to have good solvent resistance, toughness, and adhesiveness. The partially cured substrate used to manufacture circuit boards must have the characteristics of minimal flow under heat and pressure to provide adequate flow and fill, resistance to cracking, and the ability to develop adequate bond strength to all adherends during cure. When used with circuit boards, adherends include thermoplastic film, copper and/or treated copper for bond improvements, and other resinous plastic substrates used in the circuit board. When used in printed circuit boards, the dielectric loss and expansion properties of the adhesives should be adjustable to closely match those of the substrate. U.S. Patent No. 3,900,662 to Yuan discloses an acrylic based adhesive, but such adhesives have relatively low glass transition temperatures and high expansion values at soldering temperatures. Thermosetting poly(arylene ether) resin compositions have a number of the desired properties, but lower molecular weight poly(arylene ether)s useful for impregnating reinforcement materials are brittle in partially cured form while higher molecular weight poly(arylene ether)s have difficulty forming films at room temperature due to high viscosity and tend to be brittle in partially cured form. U.S. Patent No. 4,853,423 to Walles, for example, discloses poly(arylene ether) resins and polyepoxy resins together with novolac resins for circuit boards. More recent patents directed to poly(arylene ether) resins, for example U.S. Patent No. 5,834,565 to Tracy et al., disclose use of poly(arylene ether) polymers in circuit board substrate materials having number average molecular weights of less than about 3,000. Commercially available poly(arylene ether) resins have number average molecular weights in the range of about 15,000 to about 25,000. Although cured neat films of these compositions are tough, the partially cured or B-stage product tends to be brittle. There accordingly remains a need in the art for improved, less brittle thermosetting adhesives for use with thermoplastic compositions.

BRIEF SUMMARY OF THE INVENTION

In the most generic form, the invention may be described as a curable, thermosetting adhesive composition for use with thermoplastic adherends comprising a poly(arylene ether) resin having a number average molecular weight in the range from about 8,000 to about 13,000, a thermosetting resin, an optional toughening agent, a cure agent, and an optional plasticizer. In use, the composition may be blended, applied to the substrate to be adhered, partially cured or dried, and then fully cured joining the substrate to another adherend.

DETAILED DESCRIPTION OF THE INVENTION

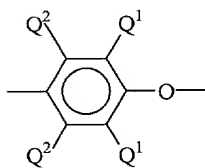
A curable, thermosetting adhesive composition comprises a poly(arylene ether) resin having a number average molecular weight in the range from about 8,000 to about 13,000, a thermosetting resin, an optional toughening agent to compatibilize the poly(arylene ether) resin and the thermosetting resin, a cure agent, and an optional plasticizer.

In an advantageous feature, the composition may be blended, applied to the substrate to be adhered, partially cured, and then fully cured joining the substrate to another adherend. Suitable bond strengths are achieved between thermoplastic substrate materials, conductive metals such as copper, and other thermoset base circuit board materials.

This invention is directed to an adhesive resin composition having better processability, exhibiting reduced B-staged (partially cured) friability, and minimal flow during lamination. Specifically, the composition, comprising a poly(arylene ether) and a thermosetting resin, is applied to a thermoplastic substrate that may have an electrically conductive metal, such as copper, on one side. Thermosetting resins are polymers that form a three-dimensional cross-linked network of polymer chains that cannot be softened or reheated for additional use. Before they are cross-linked, thermosetting resins are fluid and must contain enough reactive functionality to form a three-dimensional

network during curing. General classes of thermosetting resins include, for example, epoxy, phenolic, alkyds, acrylate, polyester, polyimide, polyurethane, bis-maleimides, cyanate esters, vinyl, benzocyclobutene resins, and benzoxazines. These compositions may further contain various catalysts, flame retardants, and other constituents, if so desired. The thermosetting components, such as those described above, may be used either alone or in combination with one another or with another thermoplastic resin. When used to coat suitable thermoplastic reinforcing material substrates such as thermoplastic polyimide, polyetherimides, polyesteretherimides, blends of poly(arylene ether) and polyimide, and blends of poly(arylene ether) and polyester, they furnish compatible substrates, particularly applicable for preparing laminated articles suitable for such applications as printed circuit boards, rigid flex circuit boards or any other articles where good dielectric properties are desired. The compositions, before crosslinking, are soluble in organic solvents, e.g., toluene, at elevated temperature and, while a gelling composition at room temperature, become liquid with shear, which facilitates coating of a substrate at room temperature. The cured adhesive layer materials prepared from the compositions are highly solder resistant, solvent resistant, moisture resistant, and flame retardant. The cured materials also have excellent dielectric properties and dimensional stability at high temperatures.

Poly(arylene ether) resins having conventional structures can be employed in the blends, comprising a plurality of structural units of the formula (I):



(I)

wherein for each structural unit, each Q^1 is independently halogen, primary or secondary lower alkyl (e.g., alkyl containing up to 7 carbon atoms), phenyl,

haloalkyl, aminoalkyl, hydrocarboxy, or halohydrocarboxy wherein at least two carbon atoms separate the halogen and oxygen atoms; and each Q^2 is independently hydrogen, halogen, primary or secondary lower alkyl, phenyl, haloalkyl, hydrocarboxy or halohydrocarboxy as defined for Q^1 .

- 5 Examples of suitable primary lower alkyl groups are methyl, ethyl, n-propyl, n-butyl, isobutyl, n-amyl, isoamyl, 2-methylbutyl, n-hexyl, 2,3-dimethylbutyl, 2-, 3-, or 4-methylpentyl and the corresponding heptyl groups. Examples of secondary lower alkyl groups are isopropyl, sec-butyl and 3-pentyl. Preferably, any alkyl radicals are straight chain rather than branched.
- 10 Preferably, each Q^1 is alkyl or phenyl, especially C_{1-14} alkyl, and each Q^2 is hydrogen.

- Both homopolymer and copolymer poly(arylene ether) polymers are included. The preferred homopolymers are those containing 2,6-dimethylphenylene ether units. Suitable copolymers include random
- 15 copolymers containing, for example, such units in combination with 2,3,6-trimethyl-1,4-phenylene ether units or copolymers derived from copolymerization of 2,6-dimethylphenol with 2,3,6-trimethylphenol. Also included are poly(arylene ether) polymers containing moieties prepared by grafting vinyl monomers or polymers such as poly(styrene), as well as
- 20 coupled poly(arylene ether) polymers in which coupling agents such as low molecular weight polycarbonates, quinones, heterocycles and formals undergo reaction in a known manner with the hydroxy groups of two poly(arylene ether) chains to produce a higher molecular weight polymer. Poly(arylene ether) polymers of the composition further include combinations
- 25 of any of the above. Many suitable random copolymers, as well as homopolymers, are disclosed in the patent literature. Reference is made to U.S. Pat. Nos. 4,054,553, 4,092,294, 4,477,649, 4,477,651 and 4,517,341, the disclosures of which are incorporated by reference herein.

- The poly(arylene ether) resin is typically prepared by the oxidative
- 30 coupling of at least one monohydroxyaromatic compound such as 2,6-xylenol or 2,3,6-trimethylphenol. Catalyst systems are generally employed for such

coupling; they typically contain at least one heavy metal compound such as a copper, manganese or cobalt compound, usually in combination with various other materials.

Particularly useful poly(arylene ether) resins are those having terminal
 5 hydroxy groups and a number average molecular weight of about 8,000 to about 13,000, more preferably about 9,000 to about 12,000, most preferably about 10,000 to about 11,000, as determined by gel permeation chromatography. The poly(arylene ether) resin accordingly has an intrinsic viscosity (I.V.) of about 0.20 to about 0.40 deciliters per gram (dl/g), preferably
 10 about 0.28 to about 0.32 dl/g, as measured in chloroform at 25°C. Such poly(arylene ether)s may be synthesized directly or obtained by subjecting poly(arylene ether)s to redistribution. Suitable methods for redistribution are described in, for example, U.S. Patent No. 5,834,565. In one procedure, low molecular weight poly(arylene ether)s are prepared from poly(arylene ether)s
 15 typically having a number average molecular weights in the range of about 15,000 to 25,000. Such preparation of a low molecular weight poly(arylene ether) resin can be accomplished by reacting the poly(arylene ether) resin with an oxidizing agent such as a peroxide or a quinone with or without a phenol (including bisphenols). Another procedure is to obtain a low molecular
 20 weight poly(arylene ether) resin by oxidative coupling as described above to produce resins of the desired number average molecular weight which is isolated, preferably, by a direct isolation method. However, even such low molecular weight resins can optionally be functionalized with a peroxide or peroxide and a phenol to achieve even lower molecular weight.

25 Phenolics useful in the redistribution reaction described herein include all known phenol compounds, including those exemplified by formula (II):



wherein A¹ is any aromatic, mixed aliphatic aromatic hydrocarbon, heterocycle or derivative of the like, X is a hydroxy residue and n is any integer from 1 to
 30 about 10 and, preferably, from 1 to about 5.

In general, any peroxide would be useful in the redistribution reaction, including those described by the following formula (III):



wherein A^2 and A^3 are any aliphatic acyl, aromatic acyl group, alkyl, or mixed aliphatic aromatic hydrocarbon, hydrogen or inorganic ester moiety or derivatives of the like. Typical peroxides include without limitation: 1) diacyl peroxides such as dibenzoyl peroxide, 4,4'-di-t-butylbenzoyl peroxide or other aryl substituted derivatives, dilauryl peroxide, acetyl benzoyl peroxide, acetyl cyclohexylsulfonyl peroxide and diphthaloyl peroxide; 2) peroxydicarbonates such as dicetylperoxydicarbonate; 3) peroxyacids such as perbenzoic acid, 3-chloroperbenzoic acid, 4-nitroperbenzoic acid, and other substituted derivatives of perbenzoic acid, peroxyacetic acid, peroxypropanoic acid, peroxybutanoic acid, peroxynonanoic acid, peroxydodecanoic acid, diperoxyglutaric acid, diperoxyadipic acid, diperoxyoctanedioic acid, diperoxynonanedioic acid, diperoxydecanedioic acid, diperoxydodecandioic acid, monoperoxyphthalic acid, as well as the inorganic acids such as peroxysulfuric acid, peroxydisulfuric acid, peroxyphosphoric acid, peroxydiphosphoric acid and their corresponding salts; and 4) peroxycarboxylic esters such as t-butyl performate, t-butyl peracetate, t-butyl peroxyisobutyrate, t-butyl perbenzoate, cumyl perbenzoate, t-butyl peroxynonanoate, t-butyl monoperoxymaleate, t-butyl monoperoxyphthalate, di-t-butyl diperoxyadipates, and 2,5-dimethyl 2,5-bis(benzoylperoxy)hexane.

These peroxides may be used alone or in combination with or without the presence of a catalyst to induce decomposition of the peroxide and increase the rate of radical production. Other oxidizing agents known in the art include quinones such as 2,2',6,6'-tetramethyl diphenquinone (TMDQ) may also be used in the presence or absence of a phenol.

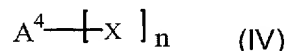
The poly(arylene ether) resin typically comprises about 5 to about 50 weight percent, preferably about 20 to about 40 weight percent, and most

preferably from about 25 to about 35 weight percent, of the total adhesive composition solids.

Adhesive compositions further comprise a thermosetting resin in its monomeric, oligomeric, or polymeric state and include reaction products with art-known curing agents and catalysts. Examples of such thermosetting resins include, for example, epoxy, phenolic, alkyds, allylic, polyester, polyimide, polyurethane, bis-maleimide, cyanate ester, vinyl, benzoxazine, and benzocyclobutene resins. Mixtures of two or more thermosetting resins may also be employed with or without added curing agents or catalysts to produce a polyphenylene ether thermosetting resin. The preferred thermosetting resin comprises an epoxide or mixtures thereof with a phenol, such as 2,2-bis(4-hydroxyphenyl)propane (bisphenol A) or bis(2-hydroxyphenyl)methane (bisphenol F), or a partial condensation product of the epoxy and bisphenols thereof. The thermosetting resin composition used herein, either as a constituent or a reactant, preferably includes at least one bisphenol polyglycidyl ether. The bisphenol polyglycidyl ether preferably employed may be halogen free or a mixture of halogen free and halogen containing polyglycidyl ethers in which bromine is the preferred halogen substituent. The total amount of bromine may be about 10 to about 30 weight percent based on the weight of the solid resin composition.

The preferred thermosetting resin comprises an epoxide or mixtures thereof with a phenol such as a bisphenol A or a partial condensation product of the epoxy and bisphenols. The thermosetting resin composition used herein, either as a constituent or a reactant, preferably includes at least one bisphenol polyglycidyl ether. The bisphenol polyglycidyl ether preferably employed may be halogen free or a mixture of halogen free and halogen containing polyglycidyl ethers in which bromine is the preferred halogen substituent. The total amount of bromine can be about 10 to about 30% by weight.

The epoxide component useful in this invention would include, in its broadest sense, any epoxy compound. Suitable epoxy compounds useful in this formulation include those of Formula (IV):

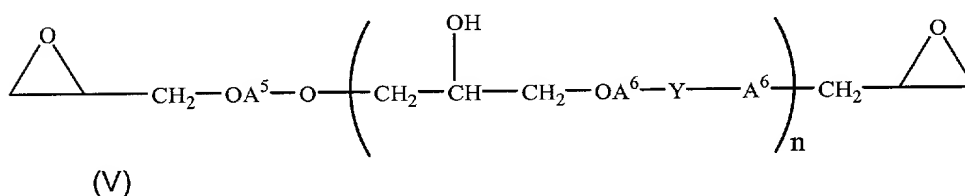


- 5 Wherein A^4 is any aromatic, aliphatic, mixed aliphatic aromatic hydrocarbon, heterocycle or derivative of the like; X is an epoxy containing residue; and n is any integer, preferably from 1 to 100.

10 When employing an epoxide, it is preferred that the epoxide comprise at least two epoxy compounds, one being brominated to provide flame retardancy and the other at levels sufficient to provide a total bromine content of about 10 to about 30 weight percent, based on the weight of the solid composition. Preferred epoxy compounds include those wherein n is 1 to 4, more preferably those wherein n is 2.

Typical to this family of materials are:

- 15 1) Diepoxides exemplified in general Formula (V)



- 20 wherein A^5 and A^6 are aromatic radicals and Y is a single bond or a bridging radical. The radicals A^5 and A^6 may be substituted or unsubstituted with typical groups chosen from aryl, alkyl, alkoxy, halo and the like. Y may include bridging radicals such as a alkylene, cycloalkylene, arylene, oxy, thio, sulfonyl, sulfoxy, and carbonyl.

- 25 Most common examples of compounds described by Formula (VI) include diglycidyl ethers often produced by the condensation of epichlorohydrin with a bisphenol where $n=0$. Typical of this class of

compounds are the diglycidyl ethers of 4,4'-(1-methylethylidene) diphenol, 4,4'-(1-methylethylidene)bis(2-methylphenol), 4,4'-(1-methylethylidene)bis(2,6-dimethylphenol), 4,4'-(1,1-cyclopentylidene)diphenol, 4,4'-(cyclohexylidene)diphenol, 4,4'-(1-methylethylidene)bis(2,6-dibromophenol), 4,4'-methylenediphenol, 4,4'-(1-methylethylidene)bis(2-allylphenol), 4,4'-(1-methylethylidene)bis(2-t-butyl-5-methylphenol), 4,4'-(1-methylethylidene)bis(2-t-butyl-5-methylphenol), 4,4'-(1-methylpropylidene)bis(2-t-butyl-5-ethylphenol), 4,4'-(1,4-bis(methylethylidene)phenyl)bis(2-t-butyl-5-methylphenol), 4,4'-biphenol, hydroquinone, resorcinol, and the like. Oligomeric products generated during this condensation reaction are also known and are useful. Such compounds are exemplified by the oligomeric condensation product of bisphenol A and epichlorohydrin (n=0.14) sold by Shell Corporation under the trademark EPON® as EPON® 828.

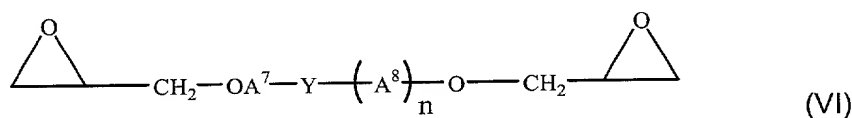
2) The reaction products of the above diepoxides with bisphenols, commonly referred to as upstaged resins. A typical example includes the condensation product of bisphenol A diglycidyl ether with tetrabromobisphenol A. The partial condensation products suitable for use may be prepared by heating the mixture of compounds, as hereinabove described, at a temperature in the range of about 50°C to about 225°C, preferably about 70°C to about 200°C, and most preferably about 100°C to about 190°C, in the presence of a catalytic amount of at least one basic reagent, such as copper (for instance lithium dimethylcuprate), amine, phosphine, or metal salt with a strong alkoxide counter ion.

The triarylphosphines, especially triphenylphosphine, are the preferred basic reagents for the bisphenol bisepoxide condensation reaction for their effectiveness at low levels, their low tendency to cause side reactions, and their harmlessness when they remain present after the reaction is complete. They are usually employed in the amount of about 0.1% to about 0.5% by weight. The reaction is preferably conducted in an inert atmosphere such as nitrogen, especially when a triarylphosphine is employed as a catalyst. An

aromatic hydrocarbon solvent such as toluene, may be employed, but is not critical.

The partial condensation products may comprise about 25% to about 35% brominated compound and about 15% to about 25% epoxidized novolac, with the balance being the non-brominated biphenol polyglycidyl ether. Lower concentrations of brominated compounds or novolacs may cause an unacceptable decrease in solvent resistance and/or flame resistance. An increase in brominated compound may yield an incompatible material. The preferred proportion of brominated compound is in the range of about 28% to about 32%.

3) Multifunctional epoxides as described by Formula (VI) may also be used.



Those materials described by Formula (VI) include all epoxidized phenolic resins including epoxidized novolacs and resols. A^7 and A^8 are aromatic radicals either substituted or unsubstituted with typical substituting groups chosen from aryl, alkyl, alkoxy, halo, and the like. The value of n may range from 0 to about 500.

Most common examples of compounds described by Formula (VI) include glycidyl ethers produced by the condensation of epichlorohydrin with a phenolic resin. Examples of this class of compounds include the glycidyl ethers of phenol formaldehyde novolac, cresol formaldehyde novolac, bromophenol formaldehyde novolac, t-butylphenol formaldehyde novolac, phenolic resins derived from the condensation of phenol with a diene or mixtures of dienes, such as dicyclopentadiene or butadiene, or additionally, with a polybutadiene resin.

Other multifunctional epoxides include phloro glucinol triglycidyl ether and tetrakis(glycidoxyphenyl) ethane.

- 4) Glycidyl ethers of amines, amides, or nitrogen containing heterocycles. These materials may include triglycidylcyanurate, triglycidylisocyanurate, N,N,N'-tetraglycidylidiaminodiphenylmethane, N,N,O-triglycidyl-4-aminophenol, N,N-diglycidyl aniline, and N,N-diglycidyl hydantoin.

5) Glycidyl ethers of carboxylic acids such as diglycidyl phthalate, diglycidyl tetrahydrophthalate, diglycidyl hexahydrophthalate and diglycidyl adipate.

- 6) Homopolymers or copolymers prepared from unsaturated epoxides such as glycidyl acrylate, glycidyl methacrylate and allyl glycidyl ether. As mentioned, these materials may be used as homopolymers or copolymers obtained from mixtures of the unsaturated epoxides mentioned above or mixtures of unsaturated epoxides and other vinyl monomers known in the practice of vinyl polymerization.

7) Polysiloxanes containing epoxy functionality such as the glycidyl ether of 1,3-bis(3-hydroxypropyl)tetramethyldisiloxane.

- 8) Compounds prepared by epoxidation of alkenes, dienes or polyenes, such as phenylglycidyl ether, allylglycidyl ether, a naphthylglycidyl ether, 3,4-epoxycyclohexylmethyl 3,4-epoxycyclohexane carboxylate, vinylcyclohexene dioxide and substituted derivatives thereof. In addition, epoxidized polyenes such as polybutadiene resins or butadiene containing copolymers would be useful.

- When the epoxide is a mixture, it usually comprises about 30 to about 60 weight percent of the brominated compound and about 5 to about 20 weight percent of the epoxidized novolac (when present), with the balance being the non brominated bisphenol polyglycidyl ether.

Other useful thermosetting components comprise vinylic compounds, including triallylisocyanurate, triallylcyanurate, diallyl phthalate, diallyl isophthalate, diallyl maleate, diallyl fumarate, diethylene glycol diallyl carbonate, triallyl phosphate, ethylene glycol diallyl ether, allyl ethers of trimethylolpropane, partial allyl ethers of pentaerythritol, diallyl sebacate, allylated novolacs, allylated resol resins, and/or cyanate esters. These various thermosetting resins can be used either individually or in combination with one another.

Cyanate esters are most broadly defined by formula (VII)



wherein A^9 is any aromatic, aliphatic, mixed aliphatic aromatic hydrocarbons, heterocycles or derivatives of the like, X is a cyanate group, and n is an integer from 1 to 10 and preferably from 1 to 4. Typical of this class of compounds are those derived from the reaction of cyanogen halides with the bisphenols described above. Various examples of cyanate esters can be found in I. Hamerton, "Chemistry and Technology of the Cyanate Esters", Chapman Hall (1994).

Polyimides, including bismaleimides, include those known in the art, such as those described in D. Wilson, H. D. Stenzenberger and P.M. Hergenrother, "Polyimides", Chapman Hall (1990).

Phenolic resins include those known to the art such as those known in the art, such as those described in A. Knop and L. A. Pilato, "Phenolic Resins: Chemistry, Applications and Performance", Springer-Verlag (1985).

Benzoxazines include those known to the art, such as those described in U.S. Patent No. 5,973,144 to Ishida.

Polyurethanes include those known in the art, such as those described in S. H. Goodman, "Handbook of Thermoset Plastics", Noyes Publications (1986).

Examples of allylic, vinyl and silicone thermosetting resins include those known in the art, such as those described in S. H. Goodman, "Handbook of Thermoset Plastics", Noyes Publications (1986).

The thermosetting resin typically comprise about 50 to about 95 weight percent, preferably about 52 to about 80 weight percent, and most preferably about 55 to about 60 weight percent by weight of the resin portion of the adhesive composition solids.

A toughening agent may be included in the composition to enhance blending and crosslinking of the poly(arylene ether) and the thermosetting resin portion of the adhesive composition. The presence of toughening agents generally reduces friability of the partially cured composition. In the final blend, the functionalized poly(arylene ether)s are commonly referred to as "toughened poly(arylene ethers)s" because of the resultant improved compatibility between the poly(arylene ether)s and the other components. Accordingly, other agents that affect the compatibility of the poly(arylene ether) with the various components of the blend (e.g., the thermosetting resin) are toughening agents. Compatibility is meant to include the stabilization of gross phase separation between the components of the blend. Indicators of improved compatibilization include, for example, increased ductility and improved phase morphology stabilization. Improved compatibility of the blend components contributes to the desirable physical properties of the adhesive.

Preferred toughening agents include polyvinyl butyral resins, including copolymers of polyvinyl butyral and other vinyl monomers such as vinyl acetate, and partially hydrolyzed derivatives therefrom. An example of such a material is poly(vinyl butyral-co-polyvinyl alcohol-co-polyvinyl acetate). Of particular utility is a polyvinyl butyral resin of molecular weight 50,000 to

120,000, available from Solutia under the trademark such as BUTVAR®.

Other toughening agents include, for example, low molecular weight thermoplastic elastomers known in the art, such as styrene-butadiene-styrene (SBS) block copolymers, styrene ethylene styrene (SES) block copolymers, styrene-ethylene-butylene-styrene (SEBS) block co-polymers and functionalized butadiene-acrylonitrile copolymers such as those sold under the tradename HYCAR® Reactive Liquid Rubbers by B. F. Goodrich Company. Also included are core shell type toughening agents such as styrene-butadiene and styrene-butadiene core shell rubbers. Other useful materials may be found in W. Hofmann and C. Hanser, "Rubber Technology Handbook", Verlag Publishers (1989); and B. Ellis, "Chemistry and Technology of the Epoxy Resins", Chapman Hall (1993).

The toughening agent typically comprise about 0.5 to about 15 weight percent, preferably about 3 to about 10 weight percent, and most preferably about 4 to about 8 weight percent, of the resin portion of the adhesive composition.

A cure agent is added to the composition to effect crosslinking between the hydroxyl groups of the poly(arylene ether) and reactive thermosetting components. For the purpose of this invention, the term cure agent is meant to include curing catalysts and co-catalysts. Any known cure agent may be employed. Suitable cure agents are described, for example, in B. Ellis, "Chemistry and Technology of the Epoxy Resins", Chapman Hall (1993). Examples of cure agents include amines, anhydrides, and carboxylates, such as, for example, metal carboxylates wherein the metal is zinc, magnesium or aluminum, and the carboxylate is a C₁₋₂₄ carboxylate such as acetate, octoate, or stearate. Additional cure agents include imidazoles and arylene polyamines, which are particularly suitable when the thermosetting resin is an epoxy resin. Cure agents are typically present at about 0.1 to about 7 weight percent, preferably from about 0.1 to about 1 weight percent, based on the total weight of resin.

A plasticizer is optionally used to reduce gelation and separation of the composition in solution, as well as to promote less brittle, partially cured substrates. When used in appropriate quantities, these plasticizers reduce the glass transition temperature of the poly(arylene ether) less than 1°C per part poly(arylene ether) without severely affecting the glass transition temperature of the remaining thermosetting component(s). Suitable plasticizers are known and include, for example, resorcinol diphosphate, bisphenol-A-diphosphate and isopropylated phenol phosphate. The plasticizer may be used at about 0 to about 20 weight percent, preferably about 0.5 to about 10 weight percent, and most preferably about 1 to about 3 weight percent.

The adhesive composition may further optionally comprise various additives, for example, antioxidants, UV absorbers, stabilizers such as light stabilizers and others, lubricants, pigments, dyes, colorants, anti-static agents, flame retardants, impact modifiers, and mixtures thereof. Exemplary antioxidants include organophosphites, for example, tris(nonyl-phenyl)phosphite, tris(2,4-di-*t*-butylphenyl)phosphite, bis(2,4-di-*t*-butylphenyl)pentaerythritol diphosphite, 2,4-di-*tert*-butylphenyl phosphite, or distearyl pentaerythritol diphosphite; alkylated monophenols, polyphenols and alkylated reaction products of polyphenols with dienes, such as, for example, tetrakis[methylene(3,5-di-*tert*-butyl-4-hydroxyhydrocinnamate)]methane and 3,5-di-*tert*-butyl-4-hydroxyhydrocinnamate octadecyl; butylated reaction products of para-cresol and dicyclopentadiene; alkylated hydroquinones; hydroxylated thiodiphenyl ethers; alkylidene-bisphenols; benzyl compounds; esters of beta-(3,5-di-*tert*-butyl-4-hydroxyphenyl)propionic acid with monohydric or polyhydric alcohols; esters of beta-(5-*tert*-butyl-4-hydroxy-3-methylphenyl)-propionic acid with monohydric or polyhydric alcohols; esters of thioalkyl or thioaryl compounds, such as, for example, distearylthiopropionate, dilaurylthiopropionate, ditridecylthiodipropionate; and amides of beta-(3,5-di-*tert*-butyl-4-hydroxyphenyl)propionic acid.

Flame retardant additives include both reactive and non-reactive flame retardant additives such as tetrabromobisphenol A derivatives, including the bis(2-hydroxyethyl)ether of tetrabromobisphenol A, the bis(3-acryloyloxy-2-hydroxypropyl) ether of tetrabromobisphenol A, the bis(3-methacryloyloxy-2-hydroxypropyl) ether of tetrabromobisphenol A, the bis(3-hydroxypropyl) ether of tetrabromobisphenol A, the bis(2,3-dibromopropyl) ether of tetrabromobisphenol A, the diallyl ether of tetrabromobisphenol A, and the bis(vinylbenzyl) ether of tetrabromobisphenol A; pentabromobenzyl acrylate; dibromostyrenes; tribromostyrenes; tetrabromocyclooctanes; dibromoethyldibromocyclohexanes such as 1,2-dibromo-4-(1,2-dibromoethyl)-cyclohexane; ethylene-bis-tetrabromophthalimide; hexabromocyclododecanes; tetrabromophthalic anhydrides; brominated diphenylethers such as decabromodiphenyl ether; poly(2,6-dibromophenylene ether); and tris(2,4,6-tribromophenoxy-1,3,5-triazine; as well as phosphorus-containing additives, for example, the phosphorus-containing additives described above and those described in R. Gachter and H. Muller (eds.), P. P. Klemchuck (assoc. ed.), "Plastic Additives Handbook, 4th Edition", Hansen Publishers, (1993). Such additives are typically used in concentrations of about 12 to about 20 weight percent of the brominated additive, or about 15 to about 25 weight percent of the phosphorous-containing additive. Flame retardance may also be imparted to the compositions by the inclusion of brominated thermosetting resins, for example a brominated poly(epoxide), or a poly(arylene ether) having a phosphorous-containing moiety in its backbone.

Where used, the flame retardant is preferably present in an amount effective to achieve a V-1 rating, as determined by Underwriters Laboratories test procedure UL 94, with V-0 being even more preferred. The V-0 rating requires a flame out time (FOT) of not more than 10 seconds in any trial and a cumulative FOT of not more than 50 seconds for five samples.

Other art-known modifiers, fillers, antioxidants, UV absorbers, stabilizers, lubricants, plasticizers, pigments, dyes, colorants, anti-static

agents, and flame retardants may also be used in the present invention such as those described in R. Gachter and H. Muller (eds.), P. P. Klemchuck (assoc. ed.), "Plastic Additives Handbook, 4th Edition", Hansen Publishers, (1993).

- 5 Fillers may also be added optionally to the adhesive compositions for the purpose of adjusting dielectric constant, loss factor, and expansion characteristics to match the adherend. Suitable fillers include but are not limited to silicates, titanium dioxide, fibers, glass fibers (including continuous and chopped fibers), carbon black, graphite, calcium carbonate, talc, and
10 mica. Suitable fillers are determined by the desired end application.

- The preparation of the adhesive composition is normally achieved by merely blending the components under conditions suitable for the formation of an intimate blend. A non-reactive solvent may also be used, for example a hydrocarbon such as toluene or xylene. During mixing, the blend is preferably
15 sufficiently heated such that the components are in solution, thereby enabling intimate mixing. Such conditions include mixing components in a vessel capable of heating and shearing action to thoroughly dissolve and blend all components.

- Suitable adherends include metals, particularly conductive metals such as copper or aluminum, and resinous thermoplastic substrate materials,
20 particularly those known for use in printed circuit boards, such as polyimide, polyetherimide, and polyesteretherimide polymers, and blends of poly(arylene ether) with polyimide or polyester polymers. A number of poly(arylene ether) compositions having favorable dielectric properties and utility in circuit board
25 manufacture are known.

 In use, the adhesive compositions are blended and then applied to the adherends by known methods, including spreading or dipping as well as various known roll transfer manufacturing methods. Solvent, if present, is removed, and the adhesive composition is partially cured. Full curing

generally requires heating at a temperature greater than about 180°C, for at least about 2.5 hours. Less than full cure may be used depending on the properties of the final product desired.

In a typical process for preparing rigid metal clad materials using the poly(arylene ether) compositions, individual lamina (commonly called prepregs) are formed from a resinous binder composition. Continuous webs of reinforcement can be pre-impregnated with the solution and then dried in a vertical or horizontal treating tower or oven. Normally, the resin is partially cured or B-staged after exiting the treating tower or oven. A metal, typically copper, foil, coated with the adhesive composition, is placed on one side of the prepreg and subjected to heating under pressure to effect a bond between the metal foil and the substrate. Multiple prepregs can be used in forming a single composite board. Additionally, multi-layer printed wiring boards will have a number of interposed laminae and copper sheets. The curing cycle in the press will depend upon the nature and thickness of the laminate, the time and temperature of the cycle being those required to cure the substrate and the bonding prepreg layer. Sufficient pressure is required to effect adequate flow of the substrate resins in order to wet out woven substrates and bond adequately. The pressure must be sufficient to prevent blistering which is due to the release of gases resulting either from retained volatiles in the substrate or adhesive layers, or resulting from by products of the curing process.

In a typical process for preparing flexible metal clad boards using the present adhesive composition, individual lamina are formed from a film, for example a polyimide film such as the polyimide film available from DuPont under the trademark KAPTON®. The metallized thermoplastic film is coated with the adhesive composition on the non-metal side, and the adhesive side of the composite is placed on a pre-made circuit inner-layer, then subjected to heating under pressure to effect a bond between the thermoplastic substrate and inner layer of circuitry. The cured adhesive layers are typically between about 0.5 and about 2.5 mils thick. Multiple metallized thermoplastic with adhesive can be used in forming a single composite board. Additionally,

multi-layer printed wiring boards will have a number of interposed laminae and copper sheets. The curing cycle in the press will again depend upon the nature and thickness of the laminate, the time and temperature of the cycle being those required to cure the substrate, and the bonding adhesive layer.

- 5 Sufficient pressure is required to effect adequate flow of the adhesive and/or substrate resins in order to wet out and bond adequately. The pressure must be sufficient to prevent blistering which is due to the release of gases resulting either from retained volatiles in the substrate or adhesive layers, or resulting from by products of the curing process.

- 10 For example, the adhesive layer may be provided having thicknesses typically of about 0.0005 inch to about 0.003 inch and the adhesives may be cured at temperatures typically in the range of about 175°C to about 205°C. The particular combination of resin thicknesses, curing times and curing
15 not limited to the particular type of resin used and the particular application. Those of ordinary skill in the art will know how to select the particular curing times and temperatures required to provide the proper dielectric for the final circuit board.

- 20 The resultant circuit board is then capable of further processing such as generation of extremely thin circuit traces due to thin copper and smooth surface of the thermoplastic film. These circuit board manufacturing techniques are known in the art. The thermoplastic/adhesive composition is fully laser ablateable for drilling as it is fully organic in nature and is useful in, for instance, microvia technologies.

- 25 The invention is further illustrated by the following non-limiting examples.

EXAMPLES

The components used in the following examples are shown in Table 1 below. The poly(arylene ether) designated "PPO (0.30 I.V.)" is not

commercially available and was prepared and isolated according to known techniques for oxidative coupling of 2,6-xylenol. Similar synthetic procedures are described in, for example, U.S. Patent No. 3,306,875 to Hay.

Table 1

Trade Name	Source	Component
PPO® (0.40 I.V.)	General Electric	Poly(arylene ether), IV = 0.40, number average molecular weight = 14,200
PPO® (0.30 I.V.)	General Electric (not commercial)	Poly(arylene ether), IV = 0.30, number average molecular weight 11,000
Bisphenol A-157	Shell Chemical	Bisphenol A
75% Dibenzoyl Peroxide, water wet	Lucidol	Benzoyl peroxide
EPON® 1163T60	Shell Chemical	Brominated epoxy resin
Upstage Resin	General Electric	EPON® 828 / TBBPA copolymer
ARALDITE® EPN1138	Ciba Geigy	Epoxy Novolac
BUTVAR® B76	Solutia	Polyvinyl butryal
KRATON® D1101	Shell Chemical	SBS block co-polymer
THERM-CHEK® 705	Ferro Corp.	Zinc Octoate
ETHACURE®100	Albemarle	Aryl amine
IMICURE® EMI-24 Curing Agent	Air Products	Imidazole accelerator
--	General Electric	resorcinol diphosphate (plasticizer)
KAPTON® - 0.002"	DuPont	Polyimide film
GOULDFLEX®-0.002" PI	Gould Electronics	Metalized polyimide film

5

Poly(arylene ether)-epoxide adhesive compositions were prepared having the compositions as outlined in the Table 2. First, the poly(arylene ether) was dissolved in toluene and tetrabromobisphenol-A diglycidyl ether to produce a solution containing approximately 40% solids. The solution was heated to 90°C to 100°C, followed by addition of bisphenol A and benzoyl

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peroxide (or, with benzoyl peroxide alone) and maintained at 90°C to 100°C for approximately 90 minutes. The solution was allowed to cool and the styrene-butadiene-styrene (SBS) block co-polymer or polyvinyl butyral compatibilizer were added. The other epoxy resins (bisphenol-A diglycidyl ether/tetrabromo bisphenol-A condensation product and epoxidized novolac) were added and the amount of toluene adjusted to generate a resin solution having 50 weight percent solids. A cure agent package consisting of zinc octoate, 2-methyl-4-ethylimidazole, and diaminodiethylbenzene was then added to complete the formulation.

10 A polyimide film was coated with each of the resin solutions described. Drawdown bars (#12 - #30) were used to apply differing thicknesses of solution to the substrate. Each coated material was heated to remove solvent and partially cure the resin to produce a B-staged coated thermoplastic material. The drum side (shiny, smooth) side of one ounce copper foil
15 (supplied by Gould Electronics) was laminated to the adhesive coated thermoplastic. Typical lamination cycles were 200 pounds per square inch (psi) and 190°C for 3 hours.

On each cured composite material physical properties were determined, and results are listed in Table 2. The amount of each component
20 shown is weight % of the solid composition. All measurements were at room temperature unless otherwise noted. Z-axis expansion was measured according to IPC-TM-650 2.4.24. Peel A and Peel after solder were measured according to IPC-TM-650 2.4.8. Solder blister times were measured according to IPC-TM-650 2.4.13.1. The three hour pressure
25 cooker test was conducted according to IPC-TM-650 2.6.16.

Solder immersion samples were solid resin bars made for this purpose.

Table 2

Component	1	2	3	4	5	6
PPO® (I.V.=0.40)	31.89	30.46	30.37	30.18	--	29.15
PPO® (I.V.=0.30)	--	--	--	--	30.65	--
Diglycidyl ether of tetrabromobisphenol A	32.56	31.1	31.0	30.82	31.29	29.76
Bisphenol A	0.32	0.30	.30	0.30	--	0.29
Benzoyl peroxide	0.32	0.32	.32	0.31	--	0.30
Epoxy novolac	14.68	14.0	13.98	13.90	14.11	13.42
Epon828/TBBPA copolymer	14.68	14.0	13.98	13.90	14.11	13.42
Polyvinyl Butyral	--	4.48	4.47	4.44	4.51	--
SBS block co-polymer	--	--	--	--	--	8.58
Zinc Octoate	4.25	4.06	4.04	4.02	4.08	3.88
Diethylenediamine	0.93	0.89	0.89	0.88	0.90	0.85
2-methyl-4-ethyl-imidazole	0.36	0.35	0.35	0.35	0.35	0.33
Plasticizer	--	--	0.30	0.91	--	--
Properties						
Tg (°C)	149/197	130/185	133/192	126/165	137/196	141/177
Z-axis expansion (30-260C, %, on neat resin bar)	--	4.18	4.37	4.48	--	--
Film Thickness (in)	0.002	0.002	0.002	0.002	0.002	0.002
Peel A (lbs/in)	5.9	6.6	7.0	7.6	6.6	8.0
Peel after solder (lbs/in)	4.6	6.1	5.8	8.1	5.8	8.3
2 min. immersion in solder at 288°C	No effect	No effect	No effect	No effect	No effect	No effect
Solder blister times (sec)	--	--	80-120	126-155	100-205	181-272
3 hour pressure cooker	Pass	Pass	Pass	Pass	--	--

It should be noted that peel failure mechanism for all samples was found to be a cohesive failure between copper and adhesive rather than

adhesive and thermoplastic. High bond strengths between adhesive and thermoplastic are desirable for reliability.

While preferred embodiments have been shown and described, various modifications and substitutions may be made thereto without departing from the spirit and scope of the invention. Accordingly, it is to be understood that
5 the present invention has been described by way of illustrations and not limitation.

All cited patents and other references are incorporated herein by reference.

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continued on next page

WHAT IS CLAIMED IS:

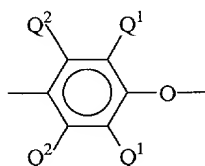
1. An adhesive formed from a composition comprising, based on 100 weight percent of the resin portion of the composition:

- 5 about 5 to about 50 weight percent of a poly(arylene ether) resin
 having a number average molecular weight of about 8,000 to about 13,000;
 about 50 to about 90 weight percent of a thermosetting resin;
 about 0.5 to about 15 weight percent of a toughening agent; and
 about 0.1 to about 7 weight percent of a cure agent.

10 2. The adhesive of Claim 1 wherein the poly(arylene ether) resin
 has a number average molecular weight of about 9,000 to about 12,000.

3. The adhesive of Claim 1 wherein the poly(arylene ether) resin
 has a number average molecular weight of about 10,000 to about 11,000.

4. The adhesive of Claim 1 wherein the poly(arylene ether) resin
 comprises a plurality of structural units of the formula (I):



(I)

15 wherein for each structural unit, each Q¹ is independently halogen, primary or
 secondary lower alkyl having up to about 7 carbon atoms, phenyl, haloalkyl,
 aminoalkyl, hydrocarbonoxy, or halohydrocarbonoxy wherein at least two
 carbon atoms separate the halogen and oxygen atoms; and each Q² is
 20 independently hydrogen, halogen, primary or secondary lower alkyl having up
 to 7 carbon atoms, phenyl, haloalkyl, hydrocarbonoxy or halohydrocarbonoxy
 wherein at least two carbon atoms separate the halogen and oxygen atoms.

5. The adhesive of Claim 4 wherein each Q¹ is an alkyl group having from 1 to 7 carbon atoms, and each Q² is hydrogen.

6. The adhesive of Claim 4 wherein the poly(arylene ether) is a homopolymer comprising 2,6-dimethylphenylene ether units, or a random copolymer comprising 2,6-dimethylphenylene ether units in combination with 2,3,6-trimethyl-1,4-phenylene ether units.

7. The adhesive of Claim 1, wherein the thermosetting resin is selected from the group consisting of cyanate esters, polyesters, epoxy, benzoxazines, benzocyclobutene resins, and mixtures comprising at least one of the foregoing thermosetting resins.

8. The adhesive of Claim 7, wherein the thermosetting resin is an epoxy resin comprising the condensation product of a bisphenol polyglycidyl ether and a bromine-substituted bisphenol.

9. The adhesive of Claim 7, wherein the epoxy is the reaction product of tetrabromobisphenol A and the diglycidyl ether of bisphenol A or bisphenol F, the reaction product having an average of at most one aliphatic hydroxy group per molecule, and the reaction product comprising about 10 to about 30 weight percent bromine as aryl substituents.

10. The adhesive of Claim 7, wherein at least one halogen-free epoxidized novolac is present.

11. The adhesive of Claim 1, wherein the thermosetting resin comprises a cyanate ester.

12. The adhesive of Claim 11, further comprising a brominated flame retardant.

13. The adhesive of Claim 1, wherein the thermosetting resin comprises a polyester.

14. The adhesive of Claim 1, wherein the thermosetting resin comprises a benzoxazine.

15. The adhesive of Claim 1, wherein the thermosetting resin comprises a benzocyclobutene resin.

5 16. The adhesive of Claim 1, wherein the thermosetting resin comprises an epoxy and a cyanate ester.

17. The adhesive of Claim 16, further comprising a brominated flame retardant.

10 18. The adhesive of Claim 1, wherein the thermosetting resin comprises a cyanate ester and an epoxy, the epoxy comprising the condensation product of a bisphenol polyglycidyl ether and a bromine-substituted bisphenol.

19. The adhesive of Claim 1, wherein the toughening agent is a polyvinyl butyral.

15 20. The adhesive of Claim 19, wherein the polyvinyl butyral is present at 1 to 15 weight percent of the total composition solids.

21. The adhesive of Claim 19, wherein the toughening agent is styrene-butadiene-styrene block co-copolymer.

20 22. The adhesive of Claim 1, further comprising a plasticizer effective for poly(arylene ether) polymers.

23. The adhesive of Claim 22, wherein the plasticizer is resorcinol diphosphate, bisphenol A diphosphate or isopropylated phenol phosphates.

25 24. The adhesive of Claim 1, further comprising one or more of modifiers, fillers, antioxidants, UV absorbers, stabilizers, lubricants, plasticizers, pigments, dyes, colorants, anti-static agents, and flame retardants.

25. The adhesive of Claim 1, comprising:
- about 20 to about 40 weight percent of the poly(arylene ether) resin;
- about 52 to about 80 weight percent of the thermosetting resin;
- 5 about 3 to about 10 weight percent of toughening agent; and
- about 0.1 to about 7 weight percent of the cure agent.
26. The adhesive of Claim 1, comprising:
- about 25 to about 35 weight percent of the poly(arylene ether) resin;
- 10 about 55 to about 65 weight percent of the thermosetting resin;
- about 4 to about 8 weight percent of the toughening agent; and
- about 0.1 to about 1 weight percent of the cure agent.
27. An adhesive formed from a composition comprising, based on 100 weight percent of the resin portion of the composition:
- 15 about 5 to about 50 weight percent of a poly(arylene ether) resin, wherein the poly(arylene ether) has a number average molecular weight of about 8,000 to about 13,000, and wherein the poly(arylene ether) is the reaction product of a higher molecular weight poly(arylene ether) with a peroxide and, optionally, a phenolic compound;
- 20 about 50 to about 90 weight percent of a thermosetting resin;
- about 0.5 to about 15 weight percent of a toughening agent; and
- about 0.1 to about 7 weight percent of a cure agent.

28. A laminate comprising:

a thermoplastic substrate;

a conductive metal foil at least partially disposed on at least one side of the substrate; and

5 an adhesive disposed between the substrate and the metal foil, wherein the adhesive is formed from a composition comprising, based on 100 weight percent of the resin portion of the composition:

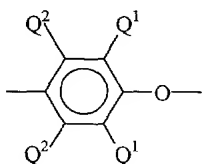
about 5 to about 50 weight percent of a poly(arylene ether) resin having a number average molecular weight of about 8,000 to about 13,000;

10 about 50 to about 90 weight percent of a thermosetting resin;

about 0.5 to about 15 weight percent of a toughening agent; and

about 0.1 to about 7 weight percent of a cure agent.

29. The laminate of claim 19, wherein the poly(arylene ether) resin comprises a plurality of structural units of the formula (I):



(I)

15 wherein for each structural unit, each Q¹ is independently halogen, primary or secondary lower alkyl having up to about 7 carbon atoms, phenyl, haloalkyl, aminoalkyl, hydrocarbonoxy, or halohydrocarbonoxy wherein at least two carbon atoms separate the halogen and oxygen atoms; and each Q² is
20 independently hydrogen, halogen, primary or secondary lower alkyl having up to 7 carbon atoms, phenyl, haloalkyl, hydrocarbonoxy or halohydrocarbonoxy wherein at least two carbon atoms separate the halogen and oxygen atoms.

30. The laminate of claim 28, wherein the thermosetting resin is selected from the group consisting of cyanate esters, polyesters, epoxy, benzoxazines, and benzocyclobutene resins.

31. The laminate of claim 30, wherein the epoxy is a condensation product of a bisphenol polyglycidyl ether and a bromine-substituted bisphenol.

32. The laminate of claim 28, wherein the toughening agent is a polyvinyl butyral or a styrene-butadiene-styrene block co-copolymer.

33. A method for forming a laminate comprising:

coating a surface of a thermoplastic or metalized thermoplastic or conductive metal foil with an adhesive comprising, based on 100 weight percent of the resin portion of the composition:

about 5 to about 50 weight percent of a poly(arylene ether) resin having a number average molecular weight of about 8,000 to about 13,000;

about 50 to about 90 weight percent of a thermosetting resin;

about 0.5 to about 15 weight percent of a toughening agent; and

about 0.1 to about 7 weight percent of a cure agent; and

applying the adhesive against a first surface of a thermoplastic substrate.

34. The method of claim 33, further comprising partially curing the adhesive prior to applying the adhesive against the substrate.

35. A method for forming an article, comprising:

coating a surface of a thermoplastic substrate with an adhesive comprising, based on 100 weight percent of the resin portion of the composition:

5 about 5 to about 50 weight percent of a poly(arylene ether) resin having a number average molecular weight of about 8,000 to about 13,000;

about 50 to about 90 weight percent of a thermosetting resin;

10 about 0.5 to about 15 weight percent of a toughening agent;

about 0.1 to about 7 weight percent of a cure agent; and

applying the adhesive against a surface of an adherend.

36. An adhesive formed from a composition comprising, based on 15 100 weight percent of the resin portion of the composition:

about 5 to about 50 weight percent of a poly(arylene ether) resin having a number average molecular weight of about 8,000 to about 13,000;

about 50 to about 90 weight percent of a thermosetting resin;

and

20 about 0.1 to about 7 weight percent of a cure agent.

ABSTRACT OF THE DISCLOSURE

32

COMBINED DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION

As a below-named inventor, we hereby declare that:

Our residences, post office address and citizenship are as stated below next to our names.

We believe we are the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: POLY(ARYLENE ETHER) ADHESIVE COMPOSITIONS, the specification of which:

☒ is attached hereto.
☐ was filed on _____ as Application Serial No. _____
and was amended on _____
(if applicable)

We hereby state that we have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We acknowledge the duty to disclose to the U.S. Patent and Trademark Office all information which is material to patentability (as defined in 37 C.F.R. § 1.56) in connection with the examination of this application.

We hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the matter provided by the first paragraph of Title 35, United States Code, Section 112, We acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, Section 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status) (patented, pending, abandoned)
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We hereby claim the benefit under Title 35, United States Code § 119(e) of any United States provisional application(s) listed below:

Provisional Application Number	Filing Date
_____	_____
_____	_____

We hereby claim foreign priority benefits under Title 35, United States Code, Section 119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

_____ Country	_____ Application No.	_____ Date of Filing	_____ Priority Claimed
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We hereby appoint; Robert E. Walter, Reg. No. 25,245; and Kenneth S. Wheelock, Reg. No. 36,340, John B. Yates, Reg. No. 39,433, Kevin E. McVeigh, Reg. No. 33,017, Frank A. Smith, Reg. No. 39,375, Michelle Bugbee, Reg. No. 42,370, all of General Electric Company, One Plastics Avenue, Pittsfield, MA 01201, Ronald E. Myrick, Reg. No. 26,315 of General Electric Company, (W3E) 3135 Easton Turnpike, Fairfield, CT 06431-0001, and Henry J. Policinski, Reg. No. 26,621 of General Electric Company, (W3D) 3135 Easton Turnpike, Fairfield, CT 06431-0001, Philmore H. Colburn II, Reg. No. 35,101 and J. Michael Buchanan, Reg. No. 44,571 of Cantor Colburn LLP, 55 Griffin Road South, Bloomfield, CT 06002, jointly and each of them severally, our attorneys or agents and attorney or agent, with full power of substitution, delegation and revocation, to prosecute this application, to make alterations and amendments therein, to receive the patent and to transact all business in the Patent and Trademark Office connected therewith. We hereby direct that all correspondence and telephone calls in connection with this application be addressed to the said:

J. Michael Buchanan, Ph.D.
at Cantor Colburn LLP, 55 Griffin Road South, Bloomfield, CT 06002
Telephone No.: (860) 286-2929

We hereby declare that all statements made herein of our own knowledge are true and that all statements made on information and belief are believed to be true; and further statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that all such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full name of sole or **first** inventor Michael J. Davis

Inventor's signature

Date

Residence 624 Ridgewood Drive Coshocton, OH 43812

Citizenship United States

Post Office Address

Full name of sole or **second** inventor James Estel Tracy

Inventor's signature

Date

Residence 225 Allison Drive, Killbuck, OH 44637

Citizenship United States

Post Office Address P.O. Box 95, 225 Allison Drive, Killbuck, OH 44637

Inventor's signature

Jeffrey H. Riding
 1000 Lane, Castleton, NY 12033

7/13/00

Date _____

Citizenship United Kingdom

Post Office Address

Inventor's signature

Date _____

Citizenship United States

Post Office Address

[illegible]

COMBINED DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION

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— was filed on _____ as Application Serial No. _____
and was amended on _____
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Filing Date

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Application No.

Date of Filing

Priority Claimed

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J. Michael Buchanan, Ph.D.
at Cantor Colburn LLP, 55 Griffin Road South, Bloomfield, CT 06002
Telephone No.: (860) 286-2929

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Full name of sole or first inventor Michael J. Davis

Inventor's signature

Date

Residence 624 Ridgewood Drive Coshocton, OH 43812

Citizenship United States

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Full name of sole or second inventor James Estel Tracy

Inventor's signature

Date

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Inventor's signature

Date _____

Citizenship United Kingdom

Post Office Address

Inventor's signature

Date _____

Citizenship United States

Post Office Address

[illegible]

COMBINED DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION

As a below-named inventor, we hereby declare that:

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We believe we are the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: POLY(ARYLENE ETHER) ADHESIVE COMPOSITIONS, the specification of which:

☒ is attached hereto.
— was filed on _____ as Application Serial No. _____
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(if applicable)

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Provisional Application Number

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Country

Application No.

Date of Filing

Priority Claimed

J. Michael Buchanan, Ph.D.
at Cantor Colburn LLP, 55 Griffin Road South, Bloomfield, CT 06002
Telephone No.: (860) 286-2929

Full name of sole or first inventor Michael J. Davis

Michael J Davis

June 28, 2000
Date

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Citizenship United States

Post Office Address

Full name of sole or second inventor James Estel Tracy

Inventor's signature

Date _____

Residence 225 Allison Drive, Killbuck, OH 44637

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Post Office Address P.O. Box 95, 225 Allison Drive, Killbuck, OH 44637

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COMBINED DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION

As a below-named inventor, we hereby declare that:

Our residences, post office address and citizenship are as stated below next to our names.

We believe we are the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: POLY(ARYLENE ETHER) ADHESIVE COMPOSITIONS, the specification of which:

X is attached hereto.
 — was filed on _____ as Application Serial No. _____
 and was amended on _____.
 (if applicable)

We hereby state that we have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We acknowledge the duty to disclose to the U.S. Patent and Trademark Office all information which is material to patentability (as defined in 37 C.F.R. § 1.56) in connection with the examination of this application.

We hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the matter provided by the first paragraph of Title 35, United States Code, Section 112, We acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, Section 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Status)
(patented, pending, abandoned)

We hereby claim the benefit under Title 35, United States Code § 119(e) of any United States provisional application(s) listed below:

Filing Date

We hereby claim foreign priority benefits under Title 35, United States Code, Section 119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

Priority Claimed

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Date _____

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8/17/2000

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